Nat Hazards (2016) 80:1709–1728
DOI 10.1007/s11069-015-2049-3

ORIGINAL PAPER

Organic and elemental carbon variation in $PM_{2.5}$ over megacity Delhi and Bhubaneswar, a semi-urban coastal site in India

Sipra Panda[1] · S. K. Sharma[2] · Parth Sarathi Mahapatra[1,3] · Upasana Panda[1] · Satyajit Rath[1] · Minakshi Mahapatra[1] · T. K. Mandal[2] · Trupti Das[1]

Received: 20 October 2014 / Accepted: 10 October 2015 / Published online: 26 October 2015

Abstract This paper presents the effect of meteorology, long-range transport, boundary layer and anthropogenic activities on the chemical composition of aerosol ($PM_{2.5}$) particularly carbonaceous aerosol (OC, EC TC) in two Indian cities, namely Delhi and Bhubaneswar. The climatological and demographical differences in the two cities have compelled the authors to compare concentrations of atmospheric organic carbon (OC) and elemental carbon (EC) in $PM_{2.5}$ at Delhi and Bhubaneswar during winter 2013 (Dec 2012 to Feb 2013). Although, Delhi is a densely populated megacity with several anthropogenic activities, Bhubaneswar is a comparatively less dense small coastal city. The percentage contribution of total carbon (TC) to $PM_{2.5}$ mass was higher as recorded at Bhubaneswar ($\sim$30.38 %) as compared to Delhi ($\sim$15 %). Average ratios of OC_{tot}/EC, K^+/OC_{tot} and K^+/EC were recorded as 1.88 $\pm$ 0.24, 0.006 $\pm$ 0.004 and 0.018 $\pm$ 0.013 at Bhubaneswar, respectively, whereas in Delhi, respective average ratios of OC_{tot}/EC, K^+/OC_{tot} and K^+/EC were recorded as 1.37 $\pm$ 0.16, 0.230 $\pm$ 0.066 and 0.321 $\pm$ 0.122. OC_{tot}/EC, K^+/OC_{tot}, K^+/EC ratios and eight carbon fraction analysis of $PM_{2.5}$ mass revealed the dominant contribution of fossil fuel specifically from coal combustion at Bhubaneswar, whereas vehicular exhaust, fossil fuel combustion along with biomass burning and road dust were the main sources of emission at Delhi. Long-range transport and prevailing meteorology had a major impact on the respective pollutants at Bhubaneswar, and OC_{tot} and EC of $PM_{2.5}$ mass over Delhi were believed to have originated from local sources due to shallow boundary layer, stable meteorology and high anthropogenic activities during the observation period. Besides, secondary organic carbon (OC_{sec}) contributed 15.76 $\pm$ 8.41 and 14.65 $\pm$ 7.46 % to OC_{tot} concentration of Bhubaneswar and Delhi, respectively.

✉ Trupti Das
trupti.sreyas@gmail.com; truptidas@immt.res.in

[1] CSIR-Institute of Minerals and Materials Technology, Bhubaneswar 751 013, India

[2] CSIR-National Physical Laboratory, Dr. K S Krishnan Road, New Delhi 110 012, India

[3] Academy of Scientific and Innovative Research (AcSIR), CSIR-Institute of Minerals and Materials Technology, Bhubaneswar, India

Keywords Organic carbon · Elemental carbon · Secondary organic carbon · Fossil fuel · Biomass burning · Correlation matrix · MACCity emission inventory

1 Introduction

Carbonaceous aerosols contribute to climate change by disturbing energy budget of the Earth (Xu et al. 2013). Elemental carbon (EC) has the potential to absorb solar radiation, resulting in positive radiative forcing in the atmosphere (Xu et al. 2013). On the other hand, organic carbon (OC) causes negative radiative forcing by scattering the Sun's radiation (Jones et al. 2005). OC is potent enough to alter surface properties of aerosols and microphysical properties of clouds as well (Sjogren et al. 2007). Besides the climate impacts, OC is hazardous to health (Winquist et al. 2015). It consists of many carcinogenic and mutagenic species (Li et al. 2008) which diminish the longevity of humans (Harrison and Yin 2000).

Incomplete combustion of biofuels, fossil fuels and open biomass burning (Bond et al. 2007; Saarikoski et al. 2008) is the major anthropogenic sources of EC and OC. Except this, OC also has secondary sources such as chemical transformation of gaseous pollutants. Researchers describe South Asia as a major source of pollutants in the world today (Lawrence and Lelieveld 2010), and India being the most populous developing country of South Asia, energy consumption from coal and other biomass and fossil fuel is on rise which in turn increases the BC (black carbon) and OC concentrations (Bond et al. 2013). Black carbon or EC is emitted as particles due to incomplete combustion of fossil fuels, biofuel and biomass. Again, results derived from the Indian Ocean Experiment (INDOEX) (Ramanathan et al. 2002) revealed a thick layer of carbon-rich absorbing aerosols spreading over a large area from northern India to the intertropical convergence zone (ITCZ), known as Atmospheric Brown Cloud (ABC), which is capable enough to perturb the energy budget of India and so also South Asia. Besides, studies by Rehman et al. (2011), Bond et al. (2013) and Saud et al. (2013) also suggest the necessity of EC and OC measurement in Indo-Gangetic Plain (IGP) of India.

In the present study, focus is on contribution of OC and EC to $PM_{2.5}$, because Seinfeld and Pandis (2006) observed that in urban locations contribution of carbonaceous aerosols to $PM_{2.5}$ may be 40 %. Previous observations taken by Mahapatra et al. (2014a) and Sharma et al. (2014a) over Bhubaneswar (an eastern coastal rapidly urbanizing site in India) and Delhi (megacity in IGP), respectively, claimed high BC and particulate matter (PM) concentrations in winter as compared to any other seasons. Backward air trajectory analysis done by Norman et al. (2001) revealed that in winter, air masses reach Bhubaneswar through the IGP region and Western Asia, considered to be the most polluted and populated regions. Bhubaneswar being situated at the eastern coastal plain, near to Bay of Bengal ($\sim$60 km), pollutants from Pakistan, West Asia and IGP enter the Bay of Bengal through Bhubaneswar (Sen et al. 2014; Kulshrestha and Kumar 2014). Therefore, for the very first time, an attempt has been made to evaluate the existence of any similarities in PM, EC and OC characteristics of Bhubaneswar and Delhi during winter months of 2012–2013. This evaluation would provide some crucial information regarding possible source of pollutants for both the locations and potential effect of polluted IGP on eastern coastal plains.

2 Study area

Measurement of $PM_{2.5}$ at Bhubaneswar was taken at the roof top of CSIR-Institute of Minerals and Materials Technology (20°29′N and 85°83′E), an urban location (altitude of 45 m above sea level) situated in the eastern coastal plains of Odisha, India. Bhubaneswar being one of the fastest developing urban areas, there is sporadic growth of different industries like fertilizer, iron and steel, agro, paper and cement in the vicinity. The monthly 24-h average temperature in winter is 23 °C with stable atmospheric conditions. More details regarding the study site and regional meteorology have been suggested elsewhere (Mahapatra et al. 2014b).

$PM_{2.5}$ samples were collected at sampling site of CSIR-National Physical Laboratory (28°38′N, 77°10E, 218 m above sea level), Delhi. Surrounding the sampling site, vast traffic density ($\sim$100 m) and agricultural fields ($\sim$500 m towards southwest) are present. According to the records of Delhi statistical handbook 2012, a total number of registered vehicles in the city were 7.77 million in 2012–2013. Besides industrial emissions, vehicular emission, secondary aerosol, biomass burning and dust storms could also increase particulate matter concentrations (Sharma et al. 2014a). Detail description of air flow pattern and other meteorological conditions over the site has been described elsewhere (Sharma et al. 2014b). The monthly 24-h average temperature in winter is 15 °C at Delhi.

3 Methodology

3.1 Sampling

$PM_{2.5}$ sampling at both the locations was carried out on pre-baked (at 550 °C for 6 h in muffle furnace) quartz microfibre (QMA) filters (47 mm diameter). Sampling time was 24 h for Delhi, whereas for Bhubaneswar, it was 8 h (from 1000 IST to 1800 IST). Pre-baked filters were desiccated for 24 h before measuring the initial (before sampling) and final weight (after sampling). The desiccated filter papers were weighed using Sartorius semi-micro balance CPA225D with 0.01 mg readability to measure the initial and final weights. Fine particulate sampler (M/s. Envirotech; Model: APM 550) and low-flow air sampler (M/s. Polltech India Ltd) were used for $PM_{2.5}$ sampling at Delhi and Bhubaneswar, respectively. $PM_{2.5}$ samples were collected at a constant flow rate of 16.7 lpm through the respective samplers. Sample was collected at both the locations on different dates between 20 December 2012 and 26 February 2013. After collecting samples, filters were stored under dry condition at −20 °C in the deep freezer prior to analysis.

3.2 Analysis of OC and EC

Following USEPA methods, IMPROVE protocol with negative pyrolysis areas zeroed (Sharma et al. 2014a), $PM_{2.5}$ samples collected from the said locations were analysed for OC and EC concentrations using OC–EC carbon analyser (Model: DRI 2001A, M/s. Atmoslytic Inc., Calabasas, CA, USA) at CSIR-NPL, Delhi. According to the principle of OC–EC analyser, $\sim$0.536 cm^2 punched area of QM-A filter was heated to 140, 280, 480 and 580 °C in pure helium to have OC_1, OC_2, OC_3 and OC_4, respectively (Chow et al. 2004). Again, for EC_1, EC_2 and EC_3, the same punched area was heated to 580, 740 and

840 °C in 98 % helium and 2 % oxygen (Chow et al. 2004; Saud et al. 2012). Each filter was analysed in triplicate with several blank runs (OC–EC analyser runs without filters to eradicate the pollutants/impurities) to get the representative estimation of OC and EC mass in $PM_{2.5}$. To perform OC and EC analysis, QM-A filters have been used by several researchers (Chen et al. 2004; Zhu et al. 2010). Moreover, to remove uncertainties in OC calculations due to volatilization of particulate organic carbon and over estimation of OC by absorption of gaseous organics, blank filters were analysed. Total OC (OC_{tot}), EC and TC were calculated using the following formulae (Chow et al. 2004; Turpin and Lim 2001; Gu et al. 2010),

$$OC_{tot} = OC_1 + OC_2 + OC_3 + OC_4 + OP$$

$$EC = EC_1 + EC_2 + EC_3 - OP$$

$$TC = OC + EC$$

$$TCA = OC \times 1.6 + EC$$

where OP is the maximum pyrolysed carbon concentrations and TCA is the total carbonaceous aerosol in urban location.

3.3 Semi-empirical EC-tracer method

As suggested by (Castro et al. 1999), in brief,

$$OC_{pri} = EC \times \left(\frac{OC}{EC}\right)_{min}$$

$$\text{And } OC_{tot} = OC_{sec} + OC_{pri}$$

$$\Rightarrow OC_{sec} = OC_{tot} - OC_{pri}$$

where OC_{pri} is the primary OC concentration, OC_{tot} is the total OC measured, OC_{sec} is the secondary organic carbon (OC_{sec}) to be calculated, $\left(\frac{OC}{EC}\right)_{min}$ is the minimum OC/EC ratio observed throughout the experiment.

3.4 Analysis of K^+

One-fourth fraction of each exposed filter paper was taken in a 50-ml stoppered test tube and extracted in 15 ml of ultrapure de-ionized water (conductivity >18.2 MΩ) in an ultrasonic bath. The filter was left overnight in the prepared solution to ensure complete solubility of the ions. The soluble components were then separated using centrifugation technique (at 2500 rpm over 10 min). The supernatant solution was filtered twice using nylon and hydrophilized poly (tetrafluoroethylene) membrane filters of 0.45 and 0.2 μm pore sizes, respectively (Das et al. 2011) in order to ensure high purity of the sample solution. The filtrate was then analysed using a Dionex ion chromatograph. The analysis of K^+ present in $PM_{2.5}$ samples collected at Bhubaneswar has been carried out by Dionex ion chromatographs (Model IC-1000), consisting of two systems for analysis of both anions and cations simultaneously along with an autosampler for accuracy and repeatability of

sampling procedure. The system was fitted with appropriate guard, separation columns, suppressors and conductivity detectors. For cation analysis, CS14 columns with CSRS suppressor were used. K^+ analysis of $PM_{2.5}$ collected at sampling site of Delhi has been carried out by ion chromatograph (Model: Dionex ICS-3000, USA) with CS14 column. Calibration standards have been prepared by National Institute of Standards and Technology (NIST, USA). The analytical error (repeatability) was estimated to be 3 % based on triplicate ($n = 3$).

3.5 Meteorological data

At Bhubaneswar, meteorological data were collected through automatic weather station (AWS) of Rainwise Inc (CC-3000) installed on the roof of CSIR-IMMT at a height of 15 m from ground level. Meteorological parameters (average temperature, relative humidity and wind speed) used in this paper were daily averages of those obtained from the AWS at 15-min frequency. The meteorological parameters (temperature, RH, wind speed, wind direction and pressure, etc.) at CSIR-NPL, Delhi, were measured using sensors of meteorological tower (five stages tower of 30 m height, 100 m away from the observational site). The tower takes observations at five different layers. We use the meteorological data available at 10 m height (i.e. temperature, RH, wind speed and wind direction) during the study days over Delhi.

3.6 MACCity emission data

MACCity emission inventory is an extension of the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), a historical emissions project, and was developed as a part of two projects funded by the European Union, namely, monitoring atmospheric composition and climate (MAAC) and CityZen developed in order to provide a consistent emission inventory through 1850–2100. Initially, historical emissions data set had been developed on a decadal basis for the period 1850–2000, and later the ACCMIP and the Representation Concentration Pathways (RCP) 8.5 emission data sets have been extended on a yearly basis for the period 1960–2020 for the anthropogenic emissions and 1960–2008 for the biomass burning emissions.

4 Results and discussion

4.1 OC and EC fractions in $PM_{2.5}$

Respective mean mass concentrations of $PM_{2.5}$, OC_{tot}, EC and TC observed at Bhubaneswar were 60.72 ± 20.1 μg/m^3 (<97.87 to >36.34 μg/m^3), 11.16 ± 4.28 μg/m^3 (<20.86 to >7.66 μg/m^3), 6.00 ± 2.26 μg/m^3 (<10.97 >3.42 μg/m^3) and 17.15 ± 6.48 μg/m^3 (<31.83 to >11.52 μg/m^3). On the other hand, mean mass concentrations of $PM_{2.5}$, OC_{tot}, EC and TC observed at Delhi were 186.25 ± 47.46 μg/m^3 (<256.02 to >108.41 μg/m^3), 16.46 ± 6.61 μg/m^3 (<26.47 to >9.60 μg/m^3), 12.04 ± 4.43 μg/m^3 (<18.21 to >5.76 μg/m^3) and 28.50 ± 10.98 μg/m^3 (<44.68 >15.36 μg/m^3), respectively (Table 1). The average TCA, determined by multiplying a factor of 1.6 to OC_{tot} (Turpin and Lim 2001), was observed to be 17.15 ± 6.48 and 30.1 ± 10.18 μg/m^3 at Bhubaneswar and Delhi, respectively (Table 1).

Table 1 OC_{tot}, EC and TC variation from $PM_{2.5}$ fraction on respective sampling days at Bhubaneswar and New Delhi

Date (Bhubaneswar)	$PM_{2.5}$ ($\mu g/m^3$)	OC_{tot}/EC	OC_{tot} ($\mu g/m^3$)	EC ($\mu g/m^3$)	TC ($\mu g/m^3$)	TCA ($\mu g/m^3$)	OC_{sec} (%)	Date (Delhi)	$PM_{2.5}$ ($\mu g/m^3$)	OC_{tot}/EC	OC_{tot} ($\mu g/m^3$)	EC ($\mu g/m^3$)	TC ($\mu g/m^3$)	TCA ($\mu g/m^3$)	OC_{sec} (%)
26-12-2012	97.87	2.06	16.50	8.02	24.52	26.12	20.75	20-12-2012	164.70	1.20	14.71	12.31	27.02	28.62	–
03-01-2013	81.66	1.99	7.66	3.86	11.52	13.12	17.89	02-01-2013	212.16	1.38	18.63	13.45	32.08	33.68	12.43
08-01-2013	47.74	1.90	20.86	10.97	31.83	33.43	14.33	08-01-2013	239.36	1.43	25.84	18.13	43.98	45.58	14.91
15-01-2013	52.76	1.98	8.12	4.10	12.22	13.82	17.64	10-01-2013	171.15	1.38	14.10	10.19	24.30	25.90	12.35
22-01-2013	72.77	1.74	10.91	6.26	17.17	18.77	6.49	17-01-2013	108.41	1.67	9.60	5.76	15.36	16.96	27.25
30-01-2013	42.66	1.61	10.20	6.33	16.53	18.13	–	24-01-2013	256.02	1.45	26.47	18.21	44.68	46.28	16.58
01-02-2013	46.31	1.58	8.51	5.39	13.90	15.50	–	11-02-2013	163.34	1.27	11.83	9.33	21.16	22.76	4.38
12-02-2013	76.44	1.70	11.42	6.72	18.14	19.74	4.11	19-02-2013	174.87	1.17	10.47	8.92	19.39	20.99	–
19-02-2013	36.34	2.37	8.10	3.42	11.52	13.12	31.08								
26-02-2013	52.63	1.89	9.29	4.91	14.20	14.20	13.77								
Average	60.72	1.88	11.16	6.00	17.15	17.15	15.76	Average	186.25	1.37	16.46	12.04	28.50	30.10	14.65
SD	20.10	0.24	4.28	2.26	6.48	6.48	8.41	SD	47.46	0.16	6.61	4.43	10.98	10.98	7.46

Ram and Sarin (2010) have reported ~30 to 35 % contribution of TC in total suspended particulate (TSP) mass at urban and rural sites of northern India. In agreement to this, percentage contribution of TC in $PM_{2.5}$ at Bhubaneswar was evaluated to be ~30.38 %. However, at Delhi, the percentage of TC in $PM_{2.5}$ was ~14.97 % which is in accordance with the reports of Sharma et al. (2014a, b) where ~18.4 % contribution of TC was observed in PM_{10} mass during the year 2010. In a recent study, Mandal et al. (2014) have also reported very high annual average concentrations of OC (93.0 ± 44.7 μg/m^3), EC (27.3 ± 13.4 μg/m^3) and TC (176.1 ± 84.7 μg/m^3; ~66 % of PM_{10} mass) in PM_{10} (280.7 ± 126.1 μg/m^3) at an industrial area in Delhi. Therefore, location-specific measurements also had an effect on the percentage contribution of TC to the PM concentration. Difference in percentage contribution of TC to $PM_{2.5}$ over Delhi and Bhubaneswar might be due to different emission sources.

Due to urbanization and rapid growth of pollution sources, $PM_{2.5}$ concentrations of Bhubaneswar and Delhi were 1.01 ± 0.34 and 3.10 ± 0.79 times higher than the National Ambient Air Quality Standards (NAAQS) guideline which is 60 μg/m^3 during the winter season. Again, OC_{tot}/EC ratio over the observation sites are 1.88 ± 0.24 (Bhubaneswar) and 1.37 ± 0.16 (Delhi) as presented in Table 1. It has been observed (Table 1) that the average OC_{tot} concentrations in $PM_{2.5}$ were 11.16 and 16.46 μg/m^3 and the average EC concentrations were 6.0 and 12.04 μg/m^3 at Bhubaneswar and Delhi, respectively. Therefore, it is obvious that a high EC concentration at Delhi leads to a comparatively lower OC_{tot}/EC ratio. These observations further supported pre-dominant contribution of fossil fuel combustion along with crustal sources to the overall $PM_{2.5}$ concentration at both the study sites. Various locations in India, like Ahmedabad, Allahabad, Agra, Kanpur, Hisar, Mt. Abu and Manora Peak, revealed a high OC/EC ratios (Rengarajan et al. 2007, Rastogi and Sarin 2009; Ram and Sarin 2010; Ram et al. 2012; Pachauri et al. 2013) basically due to biomass burning sources, while the ratio at Pune (2.9 ± 0.5) and Mumbai (2.0 ± 0.3) was comparatively lower presenting fossil fuel combustion and crustal source (Venkataraman et al. 2006; Safai et al. 2014). Table 2 shows OC/EC ratios of various study area in India and their emission sources. Saarikoski et al. (2008) found an OC/EC ratio of 0.71 and 6.6 representing vehicular and biomass burning sources, respectively. However, according to Chow et al. (1996), OC/EC ratio higher than 2 indicates the presence of secondary organic carbon (OC_{sec}). The partially elevated level of OC/EC ratio at Bhubaneswar in comparison with Delhi indicates the probability of OC_{sec} formation at the former site. For the present study, sources of OC_{tot}, EC and OC_{sec} contributions are described latter.

The percentage of EC, OC_{tot} and TC in $PM_{2.5}$ varied significantly at both the sites during the period of study (Table 3). Though the average OC_1 fraction (Table 4) over the observation site in Delhi indicated substantial contribution of biomass burning to the carbonaceous aerosol mass, the average percentage contribution of OC_{tot} to $PM_{2.5}$ at Delhi was 8.65 ± 1.55 where as at Bhubaneswar it was 19.75 ± 9.34. Similarly, the average percentage contribution of EC and TC at Bhubaneswar was 10.63 ± 5.06 and 30.38 ± 14.28, respectively, where as at Delhi average percentage contribution of EC and TC was 6.32 ± 0.97 and 14.97 ± 2.40, respectively. It can be observed that the percentage contribution of OC_{tot}, EC and TC to $PM_{2.5}$, at Bhubaneswar was almost double than that of Delhi. This observation prudently indicates that the pollution sources at Delhi were myriad with carbonaceous aerosols contributing ~15 % to the total $PM_{2.5}$ concentration, whereas at Bhubaneswar, the carbonaceous aerosols contributed almost 30 % to the total $PM_{2.5}$ concentration during the study period. This observation can be justified by the two 6-day special observation periods during November 2009 and March 2010 by Perrino et al.

Table 2 OC/EC ratio over various locations of India in winter season

Study area	Sample duration	OC/EC concentration		Source	Reference
		$PM_{2.5}$	TSP		
Delhi	Dec 2013 to Feb 2014	1.37 ± 0.16		Vehicular exhaust, fossil fuel and biomass burning	Current study
Bhubaneswar	Dec 2013 to Feb 2014	1.88 ± 0.24		Vehicular exhaust, fossil fuel burning	Current study
Allahabad	Dec 04		8.1 ± 1.7	Biomass burning	Ram and Sarin (2010)
Agra	Dec 2010 to Feb 2011	7.1	7.3	Residential combustion of wood and coal	Pachauri et al. (2013)
Ahmedabad	Dec 2004		8.3	Biomass burning	Rastogi and Sarin 2009
Kanpur	Dec 2008 to Feb 2009	7.5 ± 1.9	8.2 ± 2.0	Biomass and biofuel burning	Ram et al. (2012)
Pune	Dec 2012 to Feb 2013	2.9 ± 0.5		Fossil fuel combustion, mainly vehicular emission	Safai et al. (2014)
Mumbai	Jan–Mar 1999		3.1 ± 0.5 2.0 ± 0.3	Biomass burning and crustal source	Venkataraman et al. (2002)
Hisar	Dec 04		8.27 ± 2.19	Biomass burning and secondary organic carbon	Rengarajan et al. (2007)
Mt. Abu	Dec–Mar 2005		4.8 ± 0.9	Burning of agriculture waste	Ram et al. (2008)
Manora Peak	Feb–Mar 2005		6.6 ± 0.3	Burning of agriculture waste	Ram et al. (2008)

Table 3 Percentage contribution of OC, EC and TC to $PM_{2.5}$ at Bhubaneswar and Delhi

Bhubaneswar	OC_{tot}% in $PM_{2.5}$	EC% in $PM_{2.5}$	TC% in $PM_{2.5}$	Delhi	OC_{tot}% in $PM_{2.5}$	EC% in $PM_{2.5}$	TC% in $PM_{2.5}$
26-12-12	16.86	8.20	25.05	20-12-12	8.93	7.47	16.40
3-01-13	9.38	4.73	14.11	02-01-13	8.78	6.34	15.12
8-01-13	43.70	22.97	66.67	08-01-13	10.80	7.58	18.37
15-01-13	15.38	7.77	23.16	10-01-13	8.24	5.96	14.20
22-01-13	14.99	8.60	23.59	17-01-13	8.86	5.31	14.17
30-01-13	23.90	14.84	38.74	24-01-13	10.34	7.11	17.45
1-02-13	18.38	11.64	30.02	11-02-13	7.24	5.71	12.96
12-02-13	14.94	8.79	23.73	19-02-13	5.99	5.10	11.09
19-02-13	22.28	9.42	31.70				
26-02-13	17.65	9.34	26.99				
Average	19.75	10.63	30.38	Average	8.65	6.32	14.97
SD	9.34	5.06	14.28	SD	1.55	0.97	2.40

Table 4 Percentage of eight carbon fractions over Bhubaneswar and Delhi

Bhubaneswar	OC_{tot} fractions (%)					EC fractions (%)			Delhi	OC_{tot} fractions (%)					EC fractions (%)		
Date	OC_1	OC_2	OC_3	OC_4	OP	EC_1	EC_2	EC_3	Date	OC_1	OC_2	OC_3	OC_4	OP	EC_1	EC_2	EC_3
26-12-2012	0.33	14.97	15.74	13.78	2.49	23.57	1.43	0.49	20-12-2012	22.99	28.21	34.57	11.59	9.66	96.68	1.55	0.96
03-01-2013	0.26	17.36	15.80	5.56	7.73	27.60	2.95	0.69	02-01-2013	25.78	26.71	30.23	8.73	23.97	105.41	1.12	0.78
08-01-2013	0.16	14.04	15.84	11.41	4.59	27.52	0.97	0.28	08-01-2013	28.70	27.54	30.67	5.93	26.29	108.30	1.00	0.57
15-01-2013	0.16	18.66	17.11	5.24	5.48	24.31	3.93	0.82	10-01-2013	18.48	29.10	38.40	12.31	20.50	103.88	1.56	0.91
22-01-2013	0.00	15.20	15.79	7.51	6.17	29.01	2.27	0.52	17-01-2013	1.63	49.14	41.59	32.16	4.17	78.18	1.89	1.30
30-01-2013	0.24	14.82	15.55	9.26	3.51	27.41	2.66	0.36	24-01-2013	32.95	29.79	28.72	4.95	28.94	113.61	0.96	0.58
01-02-2013	0.00	14.82	15.68	5.03	7.62	30.21	3.96	0.79	11-02-2013	16.26	30.71	35.82	17.29	15.88	105.42	1.23	0.66
12-02-2013	0.06	15.55	16.21	8.82	3.69	27.45	1.98	0.33	19-02-2013	20.37	31.31	31.46	15.32	18.72	116.10	1.50	0.88
19-02-2013	0.00	21.87	15.80	5.12	6.68	23.52	3.47	0.61									
26-02-2013	0.07	18.94	16.33	4.86	5.84	25.63	3.80	0.77									
Average	0.13	16.62	15.98	7.66	5.38	26.62	2.74	0.57	Average	20.89	31.56	33.93	13.53	18.51	103.45	1.35	0.83
SD	0.12	2.51	0.46	3.11	1.78	2.28	1.07	0.20	SD	9.51	7.27	4.48	8.65	8.40	11.83	0.32	0.24

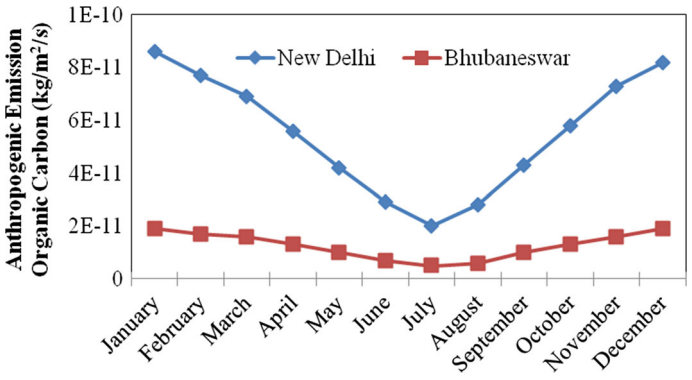

Fig. 1 Seasonal variation in anthropogenic OC emissions over geographical regions around Delhi (28.25°–29.75°N) and Bhubaneswar (19.25°–20.75°E) over the period of 4 years

(2011) who reported that combustion sources contributed to 6–7 % of the total aerosol mass, whereas the rest came from soil, inorganic secondary compounds and other organic species formed in the atmosphere.

A good correlation was observed between OC_{tot} and EC at both Bhubaneswar ($R = 0.92$) and Delhi ($R = 0.95$) indicating their common sources of emission. Further, the OC_{tot}/EC ratio at Bhubaneswar (1.88) and Delhi (1.37) indicates the dominance of motor vehicle exhaust, fossil fuel combustion and road dust during the study period. This is further supported by higher concentration of EC_1 and OC_2 fractions at both the sites (described in latter section) indicating the influence of vehicular exhaust and fossil fuel combustion.

OC concentration in Delhi reported during winter months by Srivastava et al. (2014) (38.1 ± 17.9 μg/m^3), Tiwari et al. (2013) (54 ± 39 μg/m^3), Pant et al. (2015) (104.4 μg/m^3) was significantly higher than the current study where as reports by Sharma et al. (2013, 2014a, b) are more or less similar to the OC_{tot} concentration reported here. These observations support a significant spatial variation of ambient aerosols depending on the sampling location in a megacity like Delhi.

To get a clear picture of seasonality of anthropogenically emitted OC at Delhi and Bhubaneswar, anthropogenic emissions of OC derived from MACCity emission inventory (Granier et al. 2011; Diehl et al. 2012) have been used in this study (Fig. 1). The OC emissions shown in the figure are average emission of 4 years (2010–2013) over geographical regions around Delhi (28.25°–29.75°N) and Bhubaneswar (19.25°–20.75°E). OC concentrations, derived from anthropogenic emissions, were observed more in Delhi as compared to Bhubaneswar which justifies the findings of the present study. Saud et al. (2012) observed that in IGP, dung cake is the highest contributor (1.74–4.64 g/Kg) to OC and IGP alone has 45 % contribution to total OC emissions of the country.

These results indicate the dominance of carbonaceous aerosols from fossil fuel combustion like coal, in both the locations. Therefore, further analysis was done to support this observation.

4.2 Characterization of carbon fractions

IMPROVE protocol analyses EC and OC into eight carbon fractions. These carbon fractions represent different emission sources. According to Cao et al. (2005) and Gu et al. (2010), OC_1 is the representative of biomass burning, while OC_2 of coal OC_3 and OC_4

represents road dust emissions. Similarly, EC_1 is representative of motor vehicular emissions, while EC_2 and EC_3 are mixtures of coal combustion and vehicular exhausts. In Bhubaneswar, OC_2, OC_3, OC_4, and EC_1 contribute 16.62 ± 2.51, 15.98 ± 0.46, 7.66 ± 3.11 and 26.62 ± 2.28 % (average) to TC concentrations which suggest dominance of coal combustions and vehicular emissions as sources of EC and OC (Table 4). Similarly, average variations in these carbon fractions observed in Delhi were 20.80 ± 9.51 (OC_1), 31.56 ± 7.27 (OC_2), 33.93 ± 4.48 (OC_3), 13.53 ± 8.65 (OC_4) and 103.45 ± 11.83 % (EC_1) in TC. It can be observed that at Delhi, EC_1, OC_2 and OC_3 made the maximum contribution to TC followed by OC_1 and OC_3. This indicates motor vehicle exhaust is the predominant contributed to TC followed by coal combustion, road dust and biomass burning sources at Delhi. Percentage of all the eight carbon fractions over Delhi were higher than that detected at Bhubaneswar indicating a very high pollution scenario at the national capital. Average percentage variation of OC_1 for Bhubaneswar was 0.13 ± 0.12 % (in TC) even reaching zero value on some dates indicating least contribution of biomass burning source. However, in Delhi, the period being peak winter season, maximum use of biomass for heating purposes usually contributes to a high OC_1 concentration in comparison with Bhubaneswar. Apart from biomass burning, there is also a chance of condensation of semi-volatile organic matter over the pre-existing particulate matter at lower-temperature conditions. Again, OC_2 and OC_3 abundance was relatively higher at Bhubaneswar suggesting contributions from coal combustion and road dust along with OC_{sec} production at the site (Gu et al. 2010; Cao et al. 2004). Pyrolysed carbon (OP) percentage was more in Delhi (Table 4) in comparison with Bhubaneswar. Yang and Yu (2002) observed a direct relation between OP and water-soluble organic carbon (WOCSEC). They found that WOCSEC comprises up to 13–66 % of OP.

4.3 Secondary organic carbon (OC_{sec})

Organic carbon has both primary and secondary emission sources (Ram et al. 2008; Cabada et al. 2004; Canonaco et al. 2013). Primary organic carbon (POC) is emitted from biomass and fossil fuel burning (Ram et al. 2008), while condensations of semi-volatile organic carbons (VOCs) and atmospheric transformations of biogenic and aromatic hydrocarbons contribute to OC (Rengarajan et al. 2007) collectively, known as secondary organic carbons (OC_{sec}). As direct measurement of OC_{sec} is not viable, using EC-tracer method, OC_{sec} was calculated at the locations (Turpin and Lim 2001). $\left(\frac{OC}{EC}\right)_{min}$ was taken to be 1.63 and 1.21 for Bhubaneswar and Delhi, respectively. The minimum values were determined by considering the average value of lowest three (OC/EC) ratios as suggested by Rengarajan et al. (2011).

During the sampling period, average OC_{sec} concentration at Bhubaneswar and Delhi was 1.77 and 2.57 $\mu g/m^3$, respectively, whereas for Bhubaneswar, average percentage contribution of OC_{sec} to OC_{tot} was 15.76 ± 8.41 %, and for Delhi, it was 14.65 ± 7.46 % of OC_{tot} (Table 1). Therefore, it is clear that for Bhubaneswar, OC concentrations were equally dependent on primary as well as secondary sources. Slightly higher percentage contribution of OC_{sec} at Bhubaneswar in comparison with Delhi indicates local and fresh emissions (Cao et al. 2004) from vehicular traffic, industrial emission and coal combustion from three coal-based thermal power plants surrounding the observational site of Delhi during winter (Sharma et al. 2014a). Besides the emission sources, local meteorology of a particular location plays a major role in building OC_{sec} of that area. Researchers suggest that the higher the intensity of solar radiation, the more the photochemical activity and

suitable conditions for OC_{sec} formation (Cao et al. 2004). In the present context, though the surface air temperatures of Bhubaneswar (average temperature for the study period being 22.6 ± 2.22 °C) were higher due to higher intensity of solar radiation than that of Delhi (average temperature for the study period being 12.88 ± 3.52 °C), during the sampling days, OC_{sec} concentration over the Delhi site was 1.45 times higher than that of Bhubaneswar pertaining to the myriad pollutants from various sources at the megacity Delhi. Further investigation is required to bring more clarity to such observations.

4.4 Tracing the source of EC and OC

4.4.1 Water-soluble potassium as marker of biomass burning

Various researchers (Khare and Baruah 2010; Ram and Sarin 2010; Satsangi et al. 2012) proposed water-soluble K^+ as elemental marker for biomass/wood combustion. For the first time, Andreae in 1983 proposed K^+/OC and K^+/EC ratios as indicator to emissions from biomass and fossil fuel burning, respectively. During Savanna burning (Echalar and Gaudichet 1995), K^+/OC ratio was observed to be 0.08–0.10, while it was recorded to be 0.04–0.13 for agricultural residue burning (Andreae and Merlet 2001). However, K^+/EC ratio exhibited relatively low value, 0.13 ± 0.04 at Jaduguda due to fossil fuel emissions (Ram and Sarin 2010). As suggested by Andreae (1983), K^+/EC ranging from 0.21 to 0.46 and 0.025 to 0.029 represents biomass and fossil fuel combustion, respectively. Table 5 represents ratio of K^+/EC and K^+/OC_{tot} for all measurement dates at Bhubaneswar and Delhi. Average ratio of K^+/EC and K^+/OC_{tot} over Bhubaneswar is 0.018 ± 0.013 and 0.006 ± 0.004 and at Delhi is 0.321 ± 0.122 and 0.230 ± 0.066, respectively. This observation further indicates that agricultural residue and fossil fuel combustion were the major contributors to OC and EC concentrations at Bhubaneswar. However, at Delhi, high K^+ concentrations were determined in the samples (Table 5) suggesting the role of biomass burning sources as a probable contribution to EC and OC fractions of $PM_{2.5}$. Therefore, at Delhi, biomass burning sources were supposed to be one of the contributors to carbonaceous aerosols along with fossil fuel combustion, motor vehicle exhaust and

Table 5 K^+ concentrations in $PM_{2.5}$ at Bhubaneswar and Delhi

Date (Bhubaneswar)	K^+ (μg/m^3)	K^+ % in $PM_{2.5}$	K^+/EC	K^+/OC	Date (Delhi)	K^+ (μg m^3)	K^+ % in $PM_{2.5}$	K^+/EC	K^+/OC
26-12-2012	0.087	0.089	0.005	0.015	20-12-2012	3.47	2.11	0.24	0.28
03-01-2013	0.043	0.053	0.006	0.016	02-01-2013	3.79	1.79	0.20	0.28
08-01-2013	0.004	0.008	0.000	0.001	08-01-2013	7.11	2.97	0.28	0.39
15-01-2013	0.044	0.083	0.005	0.015	10-01-2013	4.08	2.38	0.29	0.40
22-01-2013	0.158	0.217	0.014	0.036	17-01-2013	3.34	3.08	0.35	0.58
30-01-2013	0.064	0.150	0.006	0.014	24-01-2013	3.63	1.42	0.14	0.20
01-02-2013	0.024	0.052	0.003	0.006	11-02-2013	1.93	1.18	0.16	0.21
12-02-2013	0.066	0.086	0.006	0.014	19-02-2013	1.99	1.14	0.19	0.22
19-02-2013	0.112	0.308	0.014	0.046					
26-02-2013	0.038	0.072	0.004	0.011					
Average	0.064	0.112	0.006	0.018	Average	3.67	2.01	0.230	0.321
SD	0.043	0.085	0.004	0.013	SD	1.501	0.714	0.066	0.122

particles from crustal origin (Sharma et al. 2014a). Though measurement of levoglucosan a biomarker could have been a better parameter for confirming the role of biomass burning (Pant et al. 2015; Nirmalkar et al. 2015), it is out of the scope of the present study.

4.4.2 *Effect of long-range transport and wind pattern*

HYSPLIT back-trajectory model (Draxler and Rolph 2012) was used to determine the source of pollutants through long-range transport at the receptor sites (Fig. 2a). Using this on-line model, 120-h backward wind trajectories reaching Bhubaneswar and Delhi were estimated at 500 m AGL and vertical velocity mode during the observation period. Within the boundary layer, 500 m height was chosen to indicate the air mass flow. Meteorological fields for this analysis were used from the global data assimilation system (GDAS). This analysis suggests that during the observation period, air mass reaching Bhubaneswar travels through IGP and parts of western India, whereas air masses reaching Delhi travel from parts of western India, Asia, Europe and even few regions of Africa suggesting

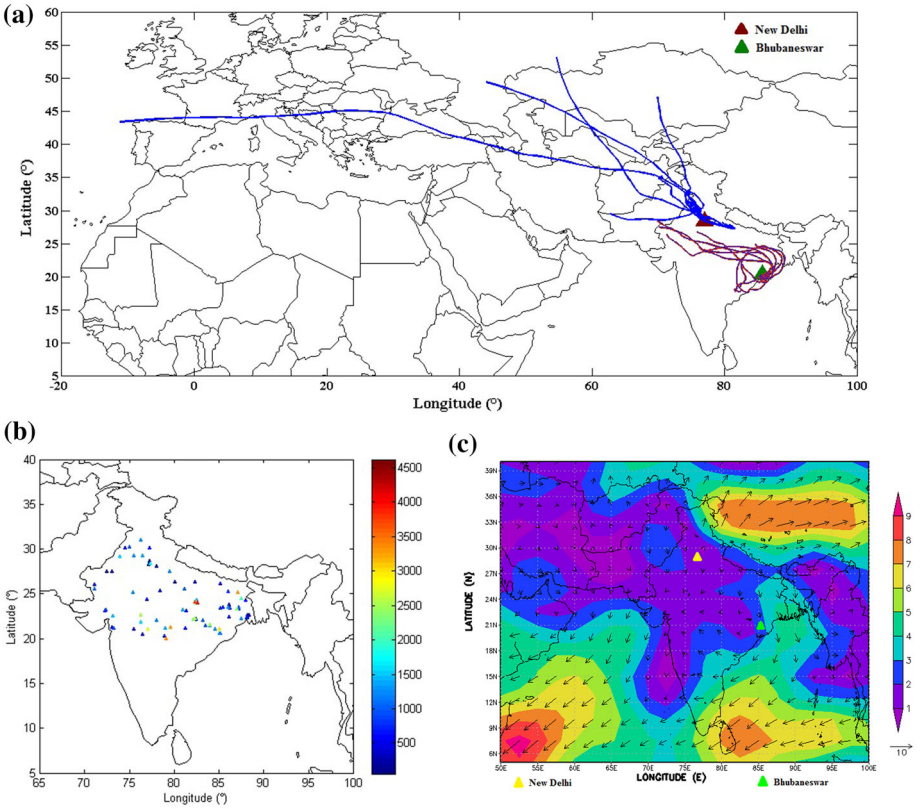

Fig. 2 **a** HYSPLIT back trajectory wind flow pattern over Bhubaneswar and Delhi. **b** Location of various coal-based thermal power plants which are considered to be the major contributors to carbonaceous aerosols at both the measurement sites. **c** Average wind speed (m/s) and wind direction during winter season over the Indian subcontinent at mean pressure level of 1000, 925 and 850 millibar using NCEP/NCAR reanalysis data

respective source regions. Analysis of eight carbon fraction, discussed earlier, suggests the presence of coal combustion as one of the major sources of carbonaceous aerosols. Hence, a map of coal-based thermal power plant spread across western, northern and eastern India along with their capacity was generated (Fig. 2b) in order to determine the role of such industries and their contribution to regional pollution through long-range transport. From the figure, it can be observed that a number of such power plants are located at a close proximity to both the observation sites. This indicates that emissions from such coal-based thermal power plants could also impact the carbonaceous aerosol concentration at both the sites through long-range transport mechanism. Further average wind speed (m/s) and wind direction during winter season over the Indian subcontinent at mean pressure level of 1000, 925 and 850 millibar were determined using NCEP/NCAR reanalysis data (Fig. 2c). The wind circulation pattern across India during winter season indicates accumulation of pollutants from the nearby regions at both the sites and lower wind speed over Delhi in comparison with Bhubaneswar. This might aid in accumulation of pollutants over Delhi in comparison with Bhubaneswar leading to higher concentration of pollutants over Delhi. Hence, the prevailing wind pattern and long-range transport aid in aerosol transport from nearby regions to the receptor site.

4.5 Role of meteorology

Meteorology has been observed to play a vital role in variation of $PM_{2.5}$ concentration (Tai et al. 2012), EC and OC variation (Lin et al. 2009). Developing a correlation matrix is traditional way of grouping few variables concerned into a matrix form and to visualize the correlation among them (Satsangi et al. 2014). Correlation matrix for this study was developed using SPSS Statistics 20 software. The relationship between various meteorological parameters (average temperature, relative humidity and wind speed), $PM_{2.5}$, EC and OC was analysed using correlation matrix (Tables 6, 7) over both the locations. At Bhubaneswar, wind speed (WS) had a weak positive but non-significant correlation with $PM_{2.5}$ ($r = 0.200$), OC_{tot} ($r = 0.386$) and EC ($r = 0.192$). This suggests that WS might not be the predominant controlling factors and more number of measurements as well as chemical characterization of PM would give a clear picture to the existing correlation (Tai et al. 2010). However, at Delhi, wind speed had a negative correlation with $PM_{2.5}$ ($r = -0.232$), OC_{tot} ($r = -0.064$) and EC ($r = -0.050$) suggesting accumulation of pollutant and hence

Table 6 Correlation matrix with meteorological parameters and rotated component at Bhubaneswar

Correlation matrix Bhubaneswar							
	$PM_{2.5}$	OC	EC	TC	Temp	Rh	Ws
Correlation							
$PM_{2.5}$	1.000						
OC	0.219	1.000					
EC	0.171	**0.961**	1.000				
TC	0.204	**0.995**	**0.983**	1.000			
Temp	−0.223	−0.650	−0.634	−0.650	1.000		
RH	0.208	0.027	−0.068	−0.006	0.009	1.000	
WS	0.200	0.386	0.192	0.322	−0.075	0.616	1.000

Numbers indicated in bold specify 95 % confidence interval

Table 7 Correlation matrix with meteorological parameters and rotated component matrix at Delhi

Correlation matrix Delhi							
	$PM_{2.5}$	OC	EC	TC	Temp	RH	Ws
Correlation							
$PM_{2.5}$	1.000						
OC	**0.926**	1.000					
EC	**0.951**	**0.977**	1.000				
TC	**0.942**	**0.996**	**0.992**	1.000			
Temp	**−0.757**	**−0.776**	**−0.771**	**−0.778**	1.000		
RH	−0.005	−0.025	−0.129	−0.067	−0.057	1.000	
Ws	−0.232	−0.064	−0.050	−0.058	−0.135	−0.542	1.000

Numbers indicated in bold specify 95 % confidence interval

a rise in concentration of $PM_{2.5}$. Temperature had a negative correlation with $PM_{2.5}$, EC and OC_{tot} at Bhubaneswar ($r = -0.23$, $r = -0.65$, $r = -0.63$) which indicates accumulation of particulates due to compressed boundary layer conditions due to low-temperature conditions. Similarly, at Delhi, $PM_{2.5}$ ($r = -0.76$), EC ($r = -0.77$) and OC_{tot} ($r = -0.78$) were negatively correlated with temperature indicating the profound role of low temperature in increasing particulate concentration. More biomass burning in homes with drop of temperature could also be a probable cause of increasing particulate matter load. At Delhi, relative humidity had a weak and non-significant negative association with $PM_{2.5}$ ($r = -0.005$) and component EC ($r = -0.13$) and OC_{tot} ($r = -0.03$); however, at Bhubaneswar, relative humidity had a non-significant but weak positive association with $PM_{2.5}$ ($r = 0.208$) and OC_{tot} ($r = 0.027$) but negative with EC ($r = -0.07$). This suggests that OC increased with higher relative humidity at Bhubaneswar.

A strong positive correlation between EC and OC_{tot} is attributed to primary emission sources of the two maintaining a fixed ratio over a particular location depending on the sources (Na et al. 2004). Though a correlation study between various meteorological parameters has been attempted with PM, EC and OC_{tot}, their quantification is beyond the scope of the manuscript as the primary focus of the paper is to undertake a comparative study of sources at both the locations. Also a fewer number of samples limit the scope of quantifying meteorological impacts on the species mentioned above. However, the results of the study are indicative that meteorology plays a vital role in variation of PM, OC_{tot} and EC, but long-term measurements would be fruitful in quantifying the same.

4.6 Role of atmospheric stability

Various studies suggest that stable atmospheric conditions play a vital role in increasing the concentration of PM and vice versa (Zhou et al. 2015; Aryal et al. 2008). Inversion conditions being an indicator of stability, an attempt was made to get an understanding of stability conditions at Bhubaneswar and Delhi on the measurement days. The same upper air data were obtained from the University of Wyoming website at 0000 GMT/0530 IST. Thermodynamic variables such as equivalent potential temperature (θ_e) were plotted against pressure (THETAPLOT diagram) where minima in θ_e were identified as characteristics of inversion (Kumar et al. 2010). THETAPLOT diagram suggested in Fig. 3a for

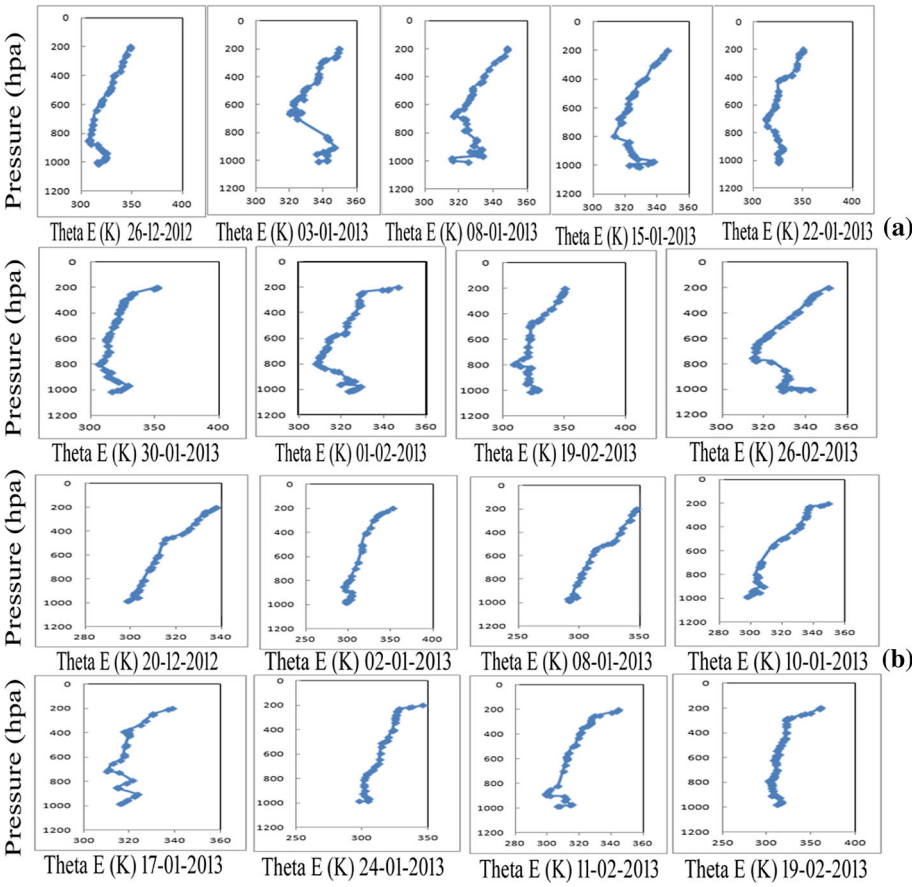

Fig. 3 Inversion graph over Bhubaneswar (19.25°–20.75°E) (**a**) and Delhi (28.25°–29.75°N) (**b**)

Bhubaneswar shows lowest equivalent potential temperature at low pressure levels ranging from 800 to 600 hPa on the sampling day, except 08 Jan 2013 (1000 hPa). Again, low pressure levels represent higher mixing height leading to vertical dispersion of pollutants. However, in Delhi, the pressure ranged between 800 and 1000 hPa (Fig. 3b) indicating a stronger inversion and more stable atmospheric condition in comparison with Bhubaneswar. Therefore, in comparison with Bhubaneswar, particulate matter trapping in air parcel under strong inversion cap could be possible which in turn increased mass of $PM_{2.5}$ as well as components EC and OC_{tot}.

5 Conclusion

Winter average concentrations of $PM_{2.5}$ at Bhubaneswar and Delhi were 60.72 ± 20.10 and 186.25 ± 47.46 μg/m^3, respectively. Percentage contributions of total carbon (TC) concentrations to $PM_{2.5}$ were higher at Bhubaneswar (~31 %) than at Delhi (~20 %) indicating almost 80 % contribution to $PM_{2.5}$ from a myriad source of pollution at the later site. The OC_{tot}/EC ratio at Bhubaneswar (1.88) and Delhi (1.37) supported the dominance

of motor vehicle exhaust, fossil fuel combustion along with road dust at both the locations. Corresponding K^+ ion contribution to $PM_{2.5}$, K^+/OC_{tot} and K^+/EC ratios of both the locations depicted that biomass burning could contribute as one of the sources to OC_{tot} and EC concentrations at Delhi, whereas fossil fuel burning could be the predominant source at Bhubaneswar. Percentage of all the eight carbon fractions were higher at Delhi indicating a very polluted atmosphere in the megacity. OC_2, OC_3 and EC_1 were the dominant species (>13 %) at Bhubaneswar among all the eight carbon fractions which indicates that coal combustion, road dust and motor vehicle exhaust are the major contributors to OC_{tot} and EC fractions. At Delhi, however, OC_1, OC_2, OC_3, OC_4, OP and EC_1 were the most abundant species (>13 %) indicating that primarily motor vehicle exhaust, coal combustion and biomass burning were contributing to the high eight carbon fractions. These observations were further supported by the wind pattern that circulated in close proximity of various major coal-based thermal power plants to both the sites. MACCity-derived anthropogenic data also indicated localized sources of OC_{tot} and EC at Delhi. On the other hand, sources of OC_{tot} and EC at Bhubaneswar were attributed to long-range transport, vehicular exhaust and fossil fuel combustion. Besides primary sources, secondary sources of organic carbon were also present at the locations; however, OC_{sec} concentration was 1.45 times more in Delhi as compared to Bhubaneswar. As this is an initial study dealing with OC_{tot} and EC concentrations in $PM_{2.5}$ over Bhubaneswar, more in-depth analysis is required to identify the exact emission sources of the carbonaceous aerosols.

Acknowledgments The authors are thankful to the Director, Institute of Minerals and Materials Technology (CSIR-IMMT), and the Head, Environment and Sustainability Department (CSIR-IMMT), for their encouragement. Authors (S.K.S., T.K.M.) are grateful to Director, CSIR-National Physical Laboratory (CSIR-NPL), for allowing to carry out this work. Financial support by ISRO-GBP (ARFI) is gratefully acknowledged. All authors acknowledge the Council of Scientific and Industrial Research (CSIR), New Delhi, for financial support under Network Project (PSC: 0112). Authors (S.R. and T.D.) acknowledge DeitY for financial support. The authors acknowledge the NOAA Air Resources Laboratory (ARL) and NASA FIRMS for the provision of the HYSPLIT model and MODIS fire events used in this publication. Authors are also thankful to Dr. Larry D. Oolman of University of Wyoming for providing upper air data of the locations.

References

Andreae MO (1983) Soot carbon and excess fine potassium: long-range transport of combustion-derived aerosols. Science 220:1148–1151

Andreae MO, Merlet P (2001) Emission of trace gases and aerosols from biomass burning. Global Biogeochem Cycles 15:955–966. doi:10.1029/2000GB001382

Aryal RK, Lee BK, Karki R, Gurung A, Kandasamy J, Pathak BK, Sharma S, Giri N (2008) Seasonal PM_{10} dynamics in Kathmandu Valley. Atmos Environ 42:8623–8633

Bond TC, Bhardwaj E, Dong R, Jogani R, Jung S, Roden C, Streets DG, Fernandes S, Trautmann N (2007) Historical emissions of black and organic carbon aerosol from energy-related combustion. Glob Biogeochem Cyc 21:1850–2000. doi:10.1029/2006GB002840

Bond TC, Doherty SJ, Fahey DW et al (2013) Bounding the role of black carbon in the climate system: a scientific assessment. J Geophys Res Atmos 118:5380–5552. doi:10.1002/jgrd.50171

Cabada JC, Pandis SN, Subramanian R et al (2004) Estimating the secondary organic aerosol contribution to $PM_{2.5}$ using the EC tracer method special issue of aerosol science and technology on findings from the fine particulate matter supersites program. Aerosol Sci Technol 38:140–155. doi:10.1080/02786820390229084

Canonaco F, Crippa M, Slowik JG et al (2013) SoFi, an IGOR-based interface for the efficient use of the generalized multilinear engine (ME-2) for the source apportionment: ME-2 application to aerosol mass spectrometer data. Atmos Meas Tech 6:3649–3661. doi:10.5194/amt-6-3649-2013

Cao J, Lee S, Ho K et al (2004) Spatial and seasonal variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China. Atmos Environ 38:4447–4456. doi:10.1016/j.atmosenv.2004.05.016

Cao JJ, Wu F, Chow JC et al (2005) Characterization and source apportionment of atmospheric organic and elemental carbon during fall and winter of 2003 in Xi'an, China. Atmos Chem Phys 5:3127–3137. doi:10.5194/acp-5-3127-2005

Castro LM, Pio CA, Harrison RM, Smith DJT (1999) Carbonaceous aerosol in urban and rural European atmospheres: estimation of secondary organic carbon concentrations. Atmos Environ 33:2771–2781. doi:10.1016/S1352-2310(98)00331-8

Chen L-WA, Chow JC, Watson JG et al (2004) Modeling reflectance and transmittance of quartz-fiber filter samples containing elemental carbon particles: implications for thermal/optical analysis. J Aerosol Sci 35:765–780. doi:10.1016/j.jaerosci.2003.12.005

Chow JC, Watson JG, Zhiqiang L, Lowenthal DH, Frazier CA, Solomon PA, Thuillier RH, Magliano K (1996) Descriptive analysis of $PM_{2.5}$ and PM_{10} at regionally representative locations during SJVAQS/AUSPEX. Atmos Environ 30:2079–2112

Chow JC, Watson JG, Chen LA et al (2004) Equivalence of elemental carbon by thermal/optical reflectance and transmittance with different temperature protocols. J Environ Sci Technol 38:4414–4422

Das N, Das R, Das SN et al (2011) Comparative studies of chemical composition of particulate matter between sea and remote location of eastern part of India. Atmos Res 99:337–343. doi:10.1016/j.atmosres.2010.11.001

Diehl T, Heil A, Chin M et al (2012) Anthropogenic, biomass burning, and volcanic emissions of black carbon, organic carbon, and SO_2 from 1980 to 2010 for hindcast model experiments. Atmos Chem Phys Discuss 12:24895–24954. doi:10.5194/acpd-12-24895-2012

Draxler RR, Rolph GD (2012) HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (http://ready.arl.noaa.gov/HYSPLIT.php). NOAA Air Resources Laboratory, Silver Spring, MD

Echalar F, Gaudichet A (1995) Aerosol emissions by tropical forest and savanna biomass burning: characteristic trace elements and fluxes. Geophys Res Lett 22:3039–3042

Granier C, Bessagnet B, Bond T et al (2011) Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period. Clim Change 109:163–190. doi:10.1007/s10584-011-0154-1

Gu J, Zhipeng B, Aixia L, Yiyang X, Weifang L, Haiyan D, Xuan Z (2010) Characterization of atmospheric organic carbon and elemental carbon of $PM_{2.5}$ and PM_{10} at Tianjin, China. Aerosol Air Qual Res 10:167–176

Harrison RM, Yin J (2000) Particulate matter in the atmosphere: which particle properties are important for its effects on health? Sci Total Environ 249:85–101

Jones GS, Jones A, Roberts DL et al (2005) Sensitivity of global-scale climate change attribution results to inclusion of fossil fuel black carbon aerosol. Lett, Geophys Res. doi:10.1029/2005GL023370

Khare P, Baruah BP (2010) Elemental characterization and source identification of $PM_{2.5}$ using multivariate analysis at the suburban site of north-east India. Atmos Res 98:148–162

Kulshrestha U, Kumar B (2014) Airmass trajectories and long range transport of pollutants: review of wet deposition scenario in South Asia. Adv Meteorol 2014:1–14. doi:10.1155/2014/596041

Kumar M, Mallik C, Kumar A, Mahanti NC, Shekh AM (2010) Evaluation of the boundary layer depth in semi-arid region of India. Dyn Atmos Ocean 49:96–107

Lawrence MG, Lelieveld J (2010) Atmospheric pollutant outflow from southern Asia: a review. Atmos Chem Phys 10:11017–11096. doi:10.5194/acp-10-11017-2010

Li H, Feng J, Sheng G et al (2008) The PCDD/F and PBDD/F pollution in the ambient atmosphere of Shanghai, China. Chemosphere 70:576–583. doi:10.1016/j.chemosphere.2007.07.001

Lin P, Hu M, Deng ZJ et al (2009) Seasonal and diurnal variations of organic carbon in $PM_{2.5}$ in Beijing and the estimation of secondary organic carbon. J Geophys Res. doi:10.1029/2008JD010902

Mahapatra PS, Panda S, Das N et al (2014a) Variation in black carbon mass concentration over an urban site in the eastern coastal plains of the Indian sub-continent. Theor Appl Climatol 117:133–147. doi:10.1007/s00704-013-0984-z

Mahapatra PS, Panda S, Walvekar PP et al (2014b) Seasonal trends, meteorological impacts, and associated health risks with atmospheric concentrations of gaseous pollutants at an Indian coastal city. Environ Sci Pollut Res 21:11418–11432. doi:10.1007/s11356-014-3078-2

Mandal P, Saud T, Sarkar R et al (2014) High seasonal variation of atmospheric C and particulate concentrations in Delhi. Environ Chem Lett, India. doi:10.1007/s10311-013-0438-y

Na K, Sawant AA, Song C, Cocker DR (2004) Primary and secondary carbonaceous species in the atmosphere of Western Riverside County, California. Atmos Environ 38:1345–1355. doi:10.1016/j.atmosenv.2003.11.023

Nirmalkar J, Deshmukh DK, Deb MK, Tsai YI, Sopajaree K (2015) Mass loading and episodic variation of molecular markers in $PM_{2.5}$ aerosols over a rural area in eastern central India. Atmos Environ 117:41–50

Norman M, Das SN et al (2001) Influence of air mass trajectories on the chemical composition of precipitation in India. Atmos Environ 35:4223–4235. doi:10.1016/S1352-2310(01)00251-5

Pachauri T, Singla V, Satsangi A et al (2013) Characterization of carbonaceous aerosols with special reference to episodic events at Agra, India. Atmos Res 128:98–110. doi:10.1016/j.atmosres.2013.03.010

Pant P, Shukla A, Kohl SD, Chow JC, Watson JG, Harrison RM (2015) Characterization of ambient $PM_{2.5}$ at a pollution hotspot in New Delhi, India and inference of sources. Atmos Environ 109:178–189

Perrino C, Tiwari S, Catrambone M, Torre SD, Ranticia E, Canepari S (2011) Chemical characterization of atmospheric PM in Delhi, India, during different periods of the year including Diwali festival. Atmos Pollut Res 2:418–427

Ram K, Sarin MM (2010) Spatio-temporal variability in atmospheric abundances of EC, OC and WOCSEC over Northern India. J Aerosol Sci 41:88–98. doi:10.1016/j.jaerosci.2009.11.004

Ram K, Sarin MM, Hegde P (2008) Atmospheric abundances of primary and secondary carbonaceous species at two high-altitude sites in India: sources and temporal variability. Atmos Environ 42(28):6785–6796

Ram K, Sarin MM, Sudheer AK, Rengarajan R (2012) Carbonaceous and secondary aerosols during wintertime fog and haze over urban sites in the Indo Gangetic Plain. Aeros Air Qual Res 12:359–370

Ramanathan V, Crutzen PJ, Mitra AP, Sikka D (2002) The Indian Ocean experiment and the Asian brown cloud. Curr Sci 83:947–955

Rastogi N, Sarin MM (2009) Quantitative chemical composition and characteristics of aerosols over western India: one-year record of temporal variability. Atmos Environ 43:3481–3488. doi:10.1016/j.atmosenv.2009.04.030

Rehman IH, Ahmed T, Praveen PS et al (2011) Black carbon emissions from biomass and fossil fuels in rural India. Atmos Chem Phys 11:7289–7299. doi:10.5194/acp-11-7289-2011

Rengarajan R, Sarin MM, Sudheer AK (2007) Carbonaceous and inorganic species in atmospheric aerosols during wintertime over urban and high-altitude sites in North India. J Geophys Res 112:D21307. doi:10.1029/2006JD008150

Rengarajan R, Sudheer AK, Sarin MM (2011) Wintertime $PM_{2.5}$ and PM_{10} carbonaceous and inorganic constituents from urban site in western India. Atmos Res 102:420–431

Saarikoski S, Timonen H, Saarnio K et al (2008) Sources of organic carbon in fine particulate matter in northern European urban air. Atmos Chem Phys 8:6281–6295. doi:10.5194/acp-8-6281-2008

Safai PD, Raju MP, Rao PSP, Pandithurai G (2014) Characterization of carbonaceous aerosols over the urban tropical location and a new approach to evaluate their climatic importance. Atmos Environ 92:493–500. doi:10.1016/j.atmosenv.2014.04.055

Satsangi A, Pachauri T, Singla V et al (2012) Organic and elemental carbon aerosols at a suburban site. Atmos Res 113:13–21. doi:10.1016/j.atmosres.2012.04.012

Satsangi PG, Yadav S, Pipal AS, Kumbhar N (2014) Characteristics of trace metals in fine ($PM_{2.5}$) and inhalable (PM_{10}) particles and its health risk assessment along with in silico approach in indoor environment of India. Atmos Environ 92:384–393. doi:10.1016/j.atmosenv.2014.04.047

Saud T, Gautam R, Mandal TK et al (2012) Emission estimates of organic and elemental carbon from household biomass fuel used over the Indo-Gangetic Plain (IGP), India. Atmos Environ 61:212–220. doi:10.1016/j.atmosenv.2012.07.030

Saud T, Saxena M, Singh DP, Saraswati Dahiya M, Sharma SK, Datta A, Gadi R, Mandal, TK (2013) Spatial variation of chemical constituents from the burning of commonly used biomass fuels in rural areas of the Indo–Gangetic Plain (IGP), India. Atmos Environ 71:158–169

Seinfeld JH, Pandis SN (2006) Atmospheric chemistry and physics: from air pollution to climate change. Wiley, London

Sen A, Mandal TK, Sharma SK et al (2014) Chemical properties of emission from biomass fuels used in the rural sector of the western region of India. Atmos Environ 99:411–424. doi:10.1016/j.atmosenv.2014.09.012

Sharma SK, Mandal TK, Saxena M, R Rohtash, Sharma A, Gautam R (2013) Source apportionment of PM10 by using positive matrix factorization at an urban site of Delhi, India. Urban Clim.doi:10.1016/j.uclim.2013.11.002

Sharma SK, Mandal TK, Saxena M et al (2014a) Source apportionment of PM_{10} by using positive matrix factorization at an urban site of Delhi, India. Urban Clim 10:656–670. doi:10.1016/j.uclim.2013.11.002

Sharma SK, Mandal TK, Saxena M et al (2014b) Variation of OC, EC, WSIC and trace metals of PM_{10} over Delhi. J Atmos Sol Terr Phys 113:10–22

Sjogren S, Gysel M, Weingartner E et al (2007) Hygroscopic growth and water uptake kinetics of two-phase aerosol particles consisting of ammonium sulfate, adipic and humic acid mixtures. J Aerosol Sci 38:157–171. doi:10.1016/j.jaerosci.2006.11.005

Srivastava AK, Bisht DS, Ram K, Tiwari S, Srivastava MK (2014) Characterization of carbonaceous aerosols over Delhi in Ganga basin: seasonal variability and possible sources. Environ Sci Pollut Res 21:8610–8619

Tai PK, Mickley LJ, Jacob DJ et al (2012) Meteorological modes of variability for fine particulate matter ($PM_{2.5}$) air quality in the United States: implications for $PM_{2.5}$ sensitivity to climate change. Atmos Chem Phys 12:3131–3145. doi:10.5194/acp-12-3131-2012

Tiwari S, Srivastava AK, Bisht DS, Safai PD, Parmita P (2013) Assessment of carbonaceous aerosol over Delhi in the Indo-Gangetic Basin: characterization, sources and temporal variability. Nat Hazards 65:1745–1764

Turpin BJ, Lim H-J (2001) Species contributions to $PM_{2.5}$ mass concentrations: revisiting common assumptions for estimating organic mass. Aerosol Sci Technol 35:602–610. doi:10.1080/02786820119445

Venkataraman C, Reddy CK, Josson S, Reddy MS (2002) Aerosol Size and Chemical Characteristics at Mumbai, India, during the INDOEX-IFP (1999). Atmos Environ 36:1979–1991

Venkataraman C, Habib G, Kadamba D et al (2006) Emissions from open biomass burning in India: integrating the inventory approach with high-resolution Moderate Resolution Imaging Spectroradiometer (MODIS) active-fire and land cover data. Global Biogeochem Cycles. doi:10.1029/2005GB002547

Winquist A, Schauer JJ, Turner JR, Klein M, Sarnat SE (2015) Impact of ambient fine particulate matter carbon measurement methods on observed associations with acute cardiorespiratory morbidity. J Eposure Sci Environ Epidemiol 25:215–221

Xu Y, Bahadur R, Zhao C, RubyLeung L (2013) Estimating the radiative forcing of carbonaceous aerosols over California based on satellite and ground observations. J Geophys Res Atmos 118:11–148. doi:10.1002/jgrd.50835

Yang HS, Yu J (2002) Uncertainties in Charring Correction in the Analysis of Elemental and Organic Carbon in Atmospheric Particles by Thermal/Optical Methods. Environ Sci Technol 36:5199–5124

Zhou B, Shen H, Huang Y, Li W, Chen H, Zhang Y, Su S, Chen Y, Lin N, Zhuo S, Zhong Q, Liu J, Li B, Tao S (2015) Daily variations of size-segregated ambient particulate matter in Beijing. Environ Pollut 197:36–42

Zhu C, Cao J, Tsai C et al (2010) The indoor and outdoor carbonaceous pollution during winter and summer in rural areas of Shaanxi. Aerosol Air Qual Res, China. doi:10.4209/aaqr.2010.04